

Written Answer

To: Director-General of the Patent Office

1. Indication of International Application: PCT/JP2004/008312

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5. Contents of answer

The applicant cannot accept the statement of the International Searching Authority and thus presents the following argument:

In regard to claims 1 to 3

(1) Document 1 discloses as follows: "An R-T-B rare earth-based sintered permanent magnet containing R_H and R_L , wherein a main phase substantially composed of $R_{L2}T_{14}B$ intermetallic compound is surrounded by an isolating phase substantially composed of a $R_{H2}T_{14}B$ intermetallic compound so that adjacent main phases are substantially isolated from each other, and a melting point lowering element M_L is added for lowering the melting point of $R_{H2}T_{14}B$ " (claim 1). In this magnet, R_H is Gd, Tb, Dy,

Ho, Er, Tm, Yb, Lu, or Y, R_L is La, Ce, Pr, Nd, Pm, Sm, or Eu, and M_L is Al, Ga, Ag, In, Bi, Sn, Pb, Zn, Ii, Sb, or Si. The permanent magnet includes the isolating phase surrounding the main phase, and essentially contains the melting point lowering element M_L , as described in the document ([0026]) that in order to realize the structure described above, the magnet of the invention contains the low-melting-point element M_L having the function to lower the melting point of the $R_{H2}T_{14}B$ intermetallic compound and having a relatively low melting point.

(2) With respect to related art, document 1 also discloses as follows: "For example, as described in Japanese Examined Patent Application Publication No. 7-78269, in order to achieve a high coercive force by a $R_2Fe_{14}B$ sintered magnet, the magnet must have a structure in which main phases composed of a tetragonal intermetallic compound are isolated from each other by a nonmagnetic R-rich phase. The nonmagnetic R-rich phase is a nonmagnetic phase having an R content of 80% or more." ([0004]).

(3) Document also discloses as follows: "For example, as shown in Figs. 1 and 2 of Japanese Examined Patent Application Publication No. 5-31807, in some conventional $R_2T_{14}B$ sintered magnets, a nonmagnetic R-rich phase appears to be present in only a portion of crystal grain boundaries. However, in such magnets, it can be confirmed by observation with a transmission electron microscope that a main phase is surrounded by a thin R-rich phase. In such conventional $R_2T_{14}B$ sintered magnets, a practical coercive force as a magnet cannot be obtained unless a main phase is surrounded by an R-rich phase." ([0014]).

(4) Furthermore, the document discloses as follows: "In order to realize a high coercive force in a conventional $R_2T_{14}B$ magnet, it is essential that main phases are isolated from each other by a nonmagnetic R-rich phase. However, the magnet of the present invention has a structure in which main phases are isolated from each other by the isolating phase functioning as a magnet instead of a nonmagnetic R-rich phase, for realizing a high coercive force and a high residual magnetic flux density. The nonmagnetic R-rich phase is preferably absent from the magnet of the present invention but may be present only in a portion (such as triple points) of crystal grain boundaries as long as each main phase is not surrounded by the R-rich phase. The nonmagnetic R-rich

phase which may be present in the magnet of the present invention is a nonmagnetic phase containing R in a larger amount than that of $R_2T_{14}B$ and generally means a phase having an R content of 80% by mass or more." ([0028]).

(5) Therefore, the invention of document 1 is characterized in that the nonmagnetic R-rich phase is absent or present only in a portion (such as triple points) of crystal grain boundaries, not having the structure essential to a conventional magnet in which main phases composed of a tetragonal intermetallic compound are isolated from each other by the nonmagnetic R-rich phase, i.e., the structure in which the main phase is surrounded by the nonmagnetic R-rich phase.

(6) The structure described in document 1 in which the isolating layer is essential, and the nonmagnetic R-rich phase is absent or slightly present is different from the structure of this invention in which a main phase is surrounded by a rare earth-rich phase (i.e., the structure described as essential to related art in document 1).

(7) The isolating phase is substantially composed of a $R_{H2}T_{14}B$ intermetallic compound and is formed by mixing a sintering raw material powder and a powder for the isolating phase without using diffusion of element M.

(8) This invention is based on the finding that penetration of element M (Pr, Dy, Tb, or Ho) into a main phase or replacement of a rare earth element in a main phase with element M is excluded as much as possible so as to enrich a rare earth-rich grain boundary phase in element M, thereby forming an element M-rich crystal grain boundary layer in which element M reacts with the rare earth-rich phase by diffusion from the surface of the magnet and thus effectively utilizing element M for improving the coercive force according to the relational expression $H_{cj} \geq 1 + 0.2 \times M$ (wherein $0.05 \leq M \leq 10$).

(9) The permanent magnet of document 1 includes a combination of a main phase and an isolating phase and does not contain an R-rich phase having a lower melting point than those of the two phases. It is thus easily supposed that it is difficult to produce a liquid phase in a sintering process and thus difficult to obtain a sintered body with a sufficiently high density. Therefore, in order to produce the permanent magnet, a low-melting-point element (nonmagnetic) such as Al, Ga, or Ag, which may decrease magnetic properties, is made essential for decreasing the melting point so that an ordinary sintering

temperature of about 1100°C is decreased to 1000°C or less (refer to claim 6).

(10) On the other hand, in order to produce the permanent magnet of this invention, a low-melting-point element need not be added to a raw material, and the method of this invention is capable of producing a sintered body having a sufficiently high density, thereby obtaining high magnetic properties under ordinary sintering conditions. Therefore, the magnet of this invention is different from the magnet of document 1 in that the magnet of this invention does not contain melting point lowering element M_L essential in decreasing the melting point of $R_{H2}T_{14}B$ of the magnet of document 1.

(11) Therefore, the invention in claims 1 to 3 of this application is distinguished from the invention described in document 1.

In regard to claims 1 to 5

(1) First, the general background of an Nd-Fe-B sintered magnetic which is a typical example of rare earth-based sintered magnets is described. It is well known that a conventional magnet includes an $Nd_2Fe_{14}B$ main phase surrounded by an Nd-rich thin phase. An $Nd_2Fe_{14}B$ sintered magnet has a coercive force of less than 1 MA/m, and use at a high temperature causes decreases in magnetic properties (maximum energy product and the amount of magnetic flux produced from the magnet), thereby producing various practical problems.

(2) It is a known fact that in order to resolve the above problem, by using the magnetic properties of $Dy_2Fe_{14}B$ and $Tb_2Fe_{14}B$ having a higher anisotropic magnetic field than that of $Nd_2Fe_{14}B$, a $(Nd,M)Fe_{14}B$ compound in which Nd is partially replaced by Dy or Tb (referred to as "M" hereinafter) is conventionally produced in a magnet, and crystal grain boundaries are partially enriched in M, thereby preventing a decrease in the magnetic properties due to an increase in coercive force at a high temperature.

(3) Also, a conventional sintered magnet is characterized in that since an Nd-Fe-B alloy powder is molded and then sintered at a high temperature of about 1100°C, element Nd of $Nd_2Fe_{14}B$ crystals is easily replaced with element M, and the residual magnetic flux density is significantly decreased although the coercive force is increased.

(4) The inventors of this invention newly found that by diffusing and penetrating

element M from the surface of a Nd-Fe-B sintered magnet and appropriately controlling the diffusion and penetration conditions, a higher coercive force can be obtained as compared with a sintered magnetic having the same element M content as that of a conventional magnet, and the residual magnetic flux density can be improved as compared with a sintered magnet having the same coercive force as that of a conventional magnet.

(5) As a result of keen consideration of the finding, it was found from analytical results that in the magnet of this invention, element M is hardly penetrated into Nd₂Fe₁₄B main phase crystals, but element M is mainly present in crystal grain boundaries between main phases. The supposed reason for this is that the diffusion rate into a Nd-rich phase which is mainly present in the original sintered magnet is higher than that into a main crystal phase.

Comparison to document 2

(1) Document 2 discloses that in a magnet produced by depositing a heavy rare earth element such as Dy or the like on the surface thereof and then thermally diffusing the element, element Dy is penetrated into the magnet. The invention of document 2 is aimed at modifying a layer damaged by machining such as cutting and grinding, and particularly, a magnet piece having a thickness of 1 mm or less and greatly degraded by machining is improved in coercive force and demagnetization curve remanence ratio. However, the object to improve the magnetic properties of a general sintered magnet itself and a specified method therefor are not described, and thus the magnetic properties such as the coercive force, residual magnetic flux density, and maximum energy product are at very low levels.

(2) In document 2, the method for depositing and diffusing element R' (= M) is a surface modifying method under substantially the same conditions as in this invention, such as the metal type, deposition method (different from three-dimensional deposition), heat treatment temperature, and the like. However, document 2 does not disclose the technical matter such as the amount of element R' distributed and the distribution state of element R' in the internal structure of the resultant magnet (i.e., the R' replacement rate of a Nd₂Fe₁₄B main phase and the amount of element R' present in a crystal grain boundary),

and the degrees of improvement of the coercive force and residual magnetic flux density due to diffusion of R'. When a R' thin film is simply deposited and diffused without being preferentially mainly diffused into crystal grain boundaries, a decrease in the residual magnetic flux density cannot be suppressed, and improvement in the maximum energy product cannot be expected.

(3) Referring to the condition represented by the relational expression, $H_{cj} \geq 1 + 0.2 \times M$ (wherein $0.05 \leq M \leq 10$), between H_{cj} (coercive force) and the element M content (% by mass) of this invention, the amounts of Tb element used in examples of document 2 and the coercive force and residual magnetic flux density values shown in Table 1 to 3 do not satisfy the above-described condition. Therefore, unlike in this invention, in document 2, element M is not effectively used by enriching a crystal grain boundary phase in element M.

(4) Therefore, document 2 neither disclose nor suggest the gist of this invention, i.e., the preferential presence of element M in crystal grain boundaries, and how to realize a high coercive force and a high residual magnetic flux density while suppressing the content of element M in a magnet.

Comparison to Document 3

(1) Document 3 discloses that in use of a magnet, high irreversible demagnetization at a high temperature is thought to be due to the lower coercive force on the surface of the magnet than that of the inside, and the irreversible demagnetization is improved by depositing element Dy or the like on the surface and diffusing it from the surface. The method of document 3 is similar to the production method of this invention in the basic material Dy, deposition of element Dy, and diffusion of element Dy into the magnet by heat treatment.

(2) However, as described above with reference to document 2, document 3 neither disclose nor suggest how to realize a high coercive force and a high residual magnetic flux density while suppressing the content of element Dy in the magnet according to purposes. Also, the Dy content and magnetic property values described in examples do not satisfy the condition of the relational expression $H_{cj} \geq 1 + 0.2 \times M$ (wherein $0.05 \leq M \leq 10$) between

H_{cj} (coercive force) and the element M content (% by mass) of this invention.

Furthermore, the data shown in tables indicates that the coercive force of the sample of each of examples 1 to 4 subjected to Dy deposition and diffusion is not at all increased as compared with the coercive force of comparative example samples. This teaches to a person skilled in the art that the deposition and diffusion do not contribute to improvement in the coercive force of the whole magnet.

(3) Therefore, document 3 does not motivate a person skilled in the art to apply the method described in document 3 to the method described in document 2 in order to increase the coercive force at a low content of element M. If the method described in document 3 is simply applied to the method described in document 2, it is impossible to obtain a permanent magnet having a specified crystal structure and magnetic properties which satisfy the relational expression $H_{cj} \geq 1 + 0.2 \times M$ (wherein $0.05 \leq M \leq 10$) which is a requirement of this invention.

(4) As described above, the invention in claims 4 and 5 of this application is similar to the methods disclosed in documents 2 and 3 in that element M is deposited and diffused. However, unlike in the inventions of documents 2 and 3, the invention in claims 4 and 5 of this application relates to a method for producing a permanent magnet having the specified crystal structure and magnetic properties of the invention defined in claims 1 to 3 of this application, not a method for simply diffusing element M. Since the permanent magnet of this invention has intensive step over documents 2 and 3, the invention relating to the production method therefor also has inventive step.